Measurement of Space Charge accumulated in Poly-dicyclopentadiene resin Film at High Temperature under High DC Stress

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Abstract—Recently, it is required for electronic device that an insulating material which shows an excellent characteristic at high temperature under high DC stress. In this study, we focused on insulating properties of Dicyclopentadiene (DCP) resin. The DCP resin shows a good thermal property and it is expected to be used as a molded insulating material alternative to the epoxy resin because of its superior moldability with low viscosity. On the other hand, polymer insulation material has a possibility of dielectric breakdown by accumulating of a space charge under a high DC stress. So, we investigated the space charge accumulation behavior in the DCP resin at high temperature under high DC stress using the PEA (Pulsed Electro-Acoustic) method.

Keywords—Dicyclopentadiene resin, space charge, epoxy resin, PEA method

I. INTRODUCTION

Recently, a miniaturization of electrical and electronic devices is progressing and it is strongly required to develop a high performance insulating material for such devices. As such an insulating material, it’s expected to show excellent properties as an insulating material at high temperature under high DC stress. In this study, we focused on the space charge accumulation in Dicyclopentadiene (DCP) resin. The DCP resin is thermosetting resin such as epoxy resin that used as a molded insulating material in many kind of electrical and electronic devices. The DCP resin shows a good thermal property and it is expected to be used alternative to the epoxy resin because of its superior moldability with low viscosity. Therefore, it is expected to be put into practical use as the electrical insulating material. On the other hand, it has been reported that a dielectric breakdown strength of certain kinds of polymeric insulating materials strongly affected by accumulation of a space charge under DC high electric field. Accumulation of the space charge sometimes causes a dielectric breakdown in the polyethylene which is the same type of hydrocarbon resin as the DCP resin [1]. Therefore, measurement of the space charge distribution can be thought effective measurement to evaluate the insulating characteristics in the DCP resin. However, there are few reports about the space charge behavior in the DCP resin at high temperature under high DC stress. In this study, we try to reveal the electrical properties of DCP resin by measuring the space charge distribution characteristics using the PEA (Pulsed Electro-Acoustic) method [2] with comparing them in epoxy resin. Furthermore, we also investigated the space charge distribution in the samples which were annealed in humid atmosphere to evaluate the effect of moisture on space charge behavior in both materials.

II. EXPERIMENTAL PROCEDURES

A. Measurement Method

Figure 1 shows a PEA measurement system at high temperature condition [3]. Temperature of the sample in silicone oil is controlled using a band heater put on the upper electrode unit as show in Fig.1.A lithium niobate (LiNbO3) crystal sensor, which enable to obtain an acoustic wave signal stably even at high temperature (30 °C -100 °C), is used as a piezo-electric sensor. A semicon-layer is used as a high voltage electrode to adjust the acoustic impedance between the aluminum electrode and the sample. In this experiment, we carried out the measurement of space charge distribution at 80 °C when high voltage of positive polarity is applied to the high voltage electrode side.

B. Sample and Measurement Condition

The measurement samples are 100 µm-thick sheets of DCP resin (PENTAM8000, RIMTEC) and DCP resin with antioxidant (BHT:dibutylhydroxytoluene) additive. We also prepared a 100 µm-thick sheet of epoxy resin sample to compare the measurement result with that obtained in DCP resin. The epoxy resin sample was manufactured by mixing a hardener (jER113, MITSUBISHI CHEMICAL) with a bisphenol A type epoxy resin (jER828, MITSUBISHI CHEMICAL). In this experiment, we investigated the space charge distribution in untreated and annealed samples. The annealed sample is previously annealed in humid atmosphere (80 °C, 80 %) for 6 hours. Measurement of the space charge distribution in each sample was carried out at 80 °C under DC stress of 150 kV/mm. In this experiment, the DC high voltages of positive polarity corresponded to the average electric field of 150 kV/mm is
applied to the each sample for 60 minutes. After voltage application, we also carried out the measurement of space charge under short circuit condition for 10 minutes.

Figure 1 PEA system for high temperature measurement

III. RESULTS AND DISCUSSION

Figure 2 shows the result of space charge measurement in the untreated samples at 80 °C under DC stress of 150 kV/mm. Figure 3 also shows the measurement results of the space charge distributions in the samples under short circuit condition after the voltage application. In these result, (A) and (B) show the results in the DCP resin and the epoxy resin respectively. In these figures, (a), (b), (c) shows the time dependent of space charge behavior described using color chart, the space charge distribution profiles, and the electric field distributions, respectively. In figures (b) and (c), red and blue lines show the results at 5 s after the start of measurement, and just before the end of measurement. As shown in Figs.2 (A) (a), (b) and (c), any remarkable space charge behavior was not observed in the untreated DCP resin under DC stress of 150 kV/mm. Consequently, any distortion of electric field by accumulated space charge was not observed in the sample. As shown in Figs.3 (A) (a) and (b), it is clear that homo space charges are observed near the anode and the cathode electrodes in the sample. By these accumulated space charges, maximum electric field of 20 kV/mm was induced in the sample under short circuit condition, as shown in Fig.3 (c). On the other hand, as shown in Figs.2 (B) (a) and (b), negative and positive homo charges were observed at the beginning of the measurement near the cathode and anode electrodes in the epoxy resin. These space charges seemed to accumulate immediately after the voltage application. However, the amount of negative charge near the cathode electrode gradually decreased, and a positive hetero charge came appear instead of the negative charge after 50 min later. Consequently, an enhancement of the electric field was observed near the cathode electrodes, which was induced by the accumulation of the hetero charge near the cathode electrode as shown in Fig.2 (B) (c). The accumulation of the positive space charge was clearly observed near the cathode electrodes under short circuit condition as shown in Fig.3 (B) (a) and (b). The maximum electric field was about 25 kV/mm induced by the space charge accumulated in the sample as shown in Fig.3 (c). Judging from the result in the untreated

![Figure 1 PEA system for high temperature measurement](image1)

![Figure 2 Space charge distributions in untreated samples at 80 °C under 150 kV/mm](image2)

![Figure 3 Space charge distributions in untreated samples at 80 °C under short circuit condition](image3)
DCP resin, small amounts of injected homo space charges from the both electrodes were observed under DC stress of 150 kV/mm in the DCP resin. On the other hand, a negative homo charge was observed near the cathode in the epoxy resin immediately after the voltage application. However, the negative charge near the cathode gradually decreased with increase of voltage application. After that, it was taken place by the accumulation of positive hetero charge. Space charge distribution in this case, it is similar to the space charge behavior at just before the dielectric breakdown in the polyimide film under high DC stress [4]. Therefore, it can be also thought that the accumulation of hetero charge in the epoxy resin might be a bad sign for the insulating property.

Figure 4 and 5 show the PEA measurement results in the annealed samples under DC stress of 150 kV/mm and the short circuit condition, respectively. In these result, (A) and (B) show the results in the DCP resin and the epoxy resin samples, respectively. In these figures, (a), (b) and (c) show the time dependents of space charge behavior described using color chart, the space charge distribution profiles, and the electric field distributions, respectively. As shown in Figs.4 (A) (a), (b) and (c), it is clearly observed that the injections of large amounts of positive and negative homo charges in the annealed DCP resin from the anode and cathode electrodes under DC stress of 150 kV/mm. Consequently, it is found that the electric field increased to about 300 kV/mm at the middle of the sample. As shown in Figs.5 (A) (a) and (b), the accumulated homo space charges were also observed near the both electrodes under short circuit condition. It is found that maximum electric field of 80 kV/mm was induced by the accumulation of the space charges in the sample under short circuit condition as shown in Fig.5 (c). On the other hand, in the case of the annealed epoxy resin, a positive hetero charge was observed near the cathode electrode just before the voltage application as shown in Fig.4 (B) (a) and (b). After that, it seems that the accumulated hetero charge moved towards the anode side with increase of the voltage application time. It is curious that the positive packet-like charge seems to move towards the anode against the electric field. While such kind of strange phenomenon has been reported as “pororoca” charge in some papers [5], it has not been recognized generally and the mechanism of its generation has not been revealed yet. Anyway, consequently, it is found that the electric field near the cathode increased to about 180 kV/mm by the accumulated positive hetero charge as shown in Fig.4 (B) (c). After that, a small amount of positive space charge was observed over all the bulk at the beginning under short circuit condition as shown in Fig.5 (B) (a) and (b). The maximum electric field induced by the space charge accumulation was about 30 kV/mm as shown in Fig.5 (c). Judging from the result in the annealed DCP resin sample, it is found that a large amount of homo charge was injected more than that in the untreated sample. An oxidation of the sample surfaces by the annealing may be assumed as one of reasons why the large amounts of the space charge injections in the bulk of the annealed DCP resin sample. Some oxidation products like as a carbonyl group is well known as a trap site for carriers and sometimes they may enhances the injection of carriers from the electrodes. Therefore, the annealing treatment on DCP resin sample may produce the injection pass into the bulk of the sample. On the other hand, since the epoxy resin is...
original including some polar groups, small amounts of homo charges may have observed even in the untreated sample. Furthermore, the change of the space charge accumulation behavior in the epoxy resin by the annealing treatment may be caused by the increase of moisture content in the bulk, because it has a high hygroscopic property. In the case of DCP resin sample, since it has a hydrophobic property, the moisture effect may not be so large. Therefore, the changes by the space charge accumulation characteristics of them may be completely different from each other.

As the results, it can be thought that space charge behavior in the DCP resin was affected by the oxidation. Therefore, we also investigated the space charge behavior in the DCP resin with antioxidant (BHT: dibutylhydroxytoluene) to reveal the effect of the oxidation on the space charge behavior in the DCP resin. Figure 6 shows the measurement results of the space charge behaviors in annealed DCP resin with antioxidant at 80 °C under DC stress of 150 kV/mm. In these results, (A) and (B) show the measurement results under DC stress and short circuit condition, respectively. In these figures, (a), (b), (c) shows the time dependent space charge behavior in color chart, the space charge distribution profiles, and the electric field distribution. As shown in Figs.6 (a), (b) and (c), a positive hetero charge was observed near the cathode electrode in the DCP resin with antioxidant under DC stress of 150 kV/mm. Consequently, an enhancement of the electric field by the accumulated hetero charge was observed near the cathode electrode. As shown in Figs.6 (B) (a), (b) and (c), the positive charge was also observed near the cathode electrodes under the short circuit condition. The maximum electric field of 30 kV/mm was induced by the accumulated space charge in the sample under short circuit condition. Judging from the result in the annealed DCP resin with antioxidant, any injected hetero charge was not observed in the DCP resin with antioxidant. Therefore, it can be thought that the reason of the increase of the injected charges in the annealed DCP resin is strongly related to the oxidation. However, it is hard to know why the positive hetero charge was accumulated in the annealed DCP resin with antioxidant. We need to investigate the effect of the positive hetero charge accumulated by adding the antioxidant on dielectric breakdown property in near the future.

IV. CONCLUSION

In this paper, we investigated the space charge distribution in the DCP resin at high temperature under high DC stress. We also carried out the space charge measurement in the epoxy resin and the DCP resin with antioxidant to compare the measurement result that obtained in DCP resin. Followings were obtained as the results.

- In an untreated DCP resin sample, any remarkable space charge accumulation was not observed in the DCP resin under DC stress of 150 kV/mm. On the other hand, a positive hetero charge was observed near the cathode in the epoxy resin.

- In an annealed DCP resin with high humid atmosphere, a large amount of injected homo space charges from electrodes were observed while it was not observed in the untreated DCP resin sample.

- In an annealed DCP resin with antioxidant any injected homo space charges from electrodes were not observed while it was observed in the DCP resin without antioxidant.

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REFERENCES


